

..... Birinci, Tutuş, Çiçekler: Evaluation of *Rhododendron luteum* and *Rhododendron*...Emre Birinci¹, Ahmet Tutuş², Mustafa Çiçekler²

Evaluation of *Rhododendron luteum* and *Rhododendron ponticum* in Pulp and Paper Production

Procjena mogućnosti uporabe biljaka *Rhododendron luteum* i *Rhododendron ponticum* kao sirovine za proizvodnju celuloze i papira

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ABSTRACT • In this study, *Rhododendron luteum* and *Rhododendron ponticum* were evaluated as raw material for pulp and paper production. 12 different sodium borohydride (NaBH_4) added cooking trials were performed for each sample and kraft method was used for pulp production. Pulp properties, such as yield, kappa number and viscosity, and physical properties, such as breaking length and burst index, were determined for each trial. Besides, the effects of active alkali and NaBH_4 on the pulp and paper properties were also examined. Optimum cooking conditions were obtained by using 18 % active alkali for NaBH_4 -free cooking experiments and 0.5 % NaBH_4 and 18 % active alkali for NaBH_4 -added cooking experiments. In NaBH_4 -added pulping condition, the screened yield, kappa number and viscosity of *R. luteum* were found to be 43.4 %, 40.1 and $949 \text{ cm}^3/\text{g}^1$, respectively. The respective values for *R. ponticum* were 41.9 %, 44.5 and $885 \text{ cm}^3/\text{g}^1$. The screened yields of *R. luteum* and *R. ponticum* increased by about 2.8 % and 5.3 %, respectively, with 5 % addition of NaBH_4 compared to NaBH_4 -free cooking experiments. Furthermore, with the addition of NaBH_4 , the kappa numbers decreased while the viscosity increased. The physical properties of the produced papers were also improved by using NaBH_4 in cooking liquor. According to the obtained results, it was found that *R. luteum* and *R. ponticum* species can be evaluated for pulp and paper production.

Keywords: *R. luteum*; *R. ponticum*; pulp; paper; NaBH_4

SAŽETAK • U ovom je radu istražena mogućnost uporabe biljaka *Rhododendron luteum* i *Rhododendron ponticum* kao sirovine za proizvodnju celuloze i papira. Za svaki uzorak provedeno je 12 različitih ispitivanja kuhanja s natrijevim borhidridom (NaBH_4), a celuloza je proizvedena kraft postupkom. Za svako ispitivanje određena su svojstva celuloze poput prinosa, kappa broja i viskoznosti, te fizička svojstva kao što su duljina lomljenja i indeks pucanja papira. Osim toga, ispitani su učinci aktivne lužine i NaBH_4 na svojstva celuloze i papira. Optimalni uvjeti kuhanja postignuti su upotrebom 18 % aktivne lužine za eksperimentalno kuhanje bez NaBH_4 i upotrebom 0,5 % NaBH_4 i 18 % aktivne lužine za eksperimentalno kuhanje s dodatkom NaBH_4 . U proizvodnji celuloze iz biljke *R. luteum* s dodatkom NaBH_4 utvrđeno je da prinos prosijavanja iznosi 43,4 %, da je kappa broj 40,1, a viskoznost

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949 cm³/g, dok su vrijednosti za celulozu dobivenu iz biljke *R. ponticum* redom 41,9 %, 44,5 i 885 cm³/g. Prinos prosijavanja biljaka *R. luteum* i *R. ponticum* uz dodatak 5 % NaBH₄ povećao se oko 2,8 % i 5,3 % u usporedbi s eksperimentalnim kuhanjem bez dodatka NaBH₄. Nadalje, uz dodatak NaBH₄ smanjuju se kappa brojevi, a viskoznosti se povećavaju. Fizika svojstva proizvedenih papira također se poboljšavaju dodavanjem NaBH₄ tekućini za kuhanje. Iz dobivenih je rezultata utvrđeno da se biljke *R. luteum* i *R. ponticum* mogu upotrebljavati u proizvodnji celuloze i papira.

Ključne riječi: *R. luteum*; *R. ponticum*; celuloza; papir; NaBH₄

1 INTRODUCTION

1. UVOD

In the second half of the twentieth century, especially with the increase of world population and the increase in demand for forest products due to rapid developments in technology, the decrease in forest areas led the forest products industry to search for raw materials as an alternative to wood (Oner and Aslan, 2002; Tutus *et al.*, 2011). Non-wood and new wood products are one of the best alternative raw materials (Kaldor, 1992; Comlekcioglu *et al.*, 2016). Due to the rapid increase in paper consumption in recent years, alternative fibrous materials have gained importance in the pulp and paper industry (Daud and Law, 2011). The search for new wood raw materials still continues in the world.

Rhododendron species, a valuable ornamental shrub, have been cultivated since the 18th century. These species belong to the heath or Ericaceae family. Rhododendron, with about 1000 different species, extends from Southwest Asia to New Guinea. Some Rhododendron species are large species with a tree-type growth habit. It is evergreen or rarely deciduous. Turkish rhododendrons grow naturally up to 3000 m above the sea level. One of the most remarkable Rhododendron species in moist forest formations covering the northern coast of Turkey is *R. luteum* and the most common Rhododendron is *R. ponticum*. Fiber length, fiber width, lumen diameter and cell wall thicknesses of *R. luteum* and *R. ponticum* are approximately 0.9-1.0 mm, 17-19 μm, 7-9 μm and 3-4 μm, respectively. Besides, these species have high holocellulose (77-80 %) and alpha cellulose (47-48 %) contents, which are important carbohydrates for pulp and paper industry (Birinci, 2008; Camlibel, 2008).

Although they appear on the northern slopes of the mountains on the Black Sea coast, there are more species in the Eastern Black Sea Region (Avci, 2004). In many studies, Rhododendron species have generally been evaluated for the production of medium density fiberboard (MDF) (Akgul *et al.*, 2012; Akgul and Camlibel, 2008; Ayrimis *et al.*, 2014). Besides, some studies have determined the chemical composition of some Rhododendron species (Birinci, 2008; Shrestha and Budhathoki, 2012).

The kraft method is the most commonly used method for obtaining pulp suitable for papermaking. Increasing the pulp yield in pulp production is very important for the pulp and paper industry in terms of cost and economy. Modifying the kraft cooking process, e.g. by adding NaBH₄, is a way of increasing the pulp

yield (Courchene, 1998; Tutus and Eroglu, 2003; Tutus and Eroglu, 2004; Tutus and Usta 2004; Hafizoglu and Deniz, 2007; Istek and Gonteki, 2009, Akgul *et al.*, 2018). The end groups of carbohydrates are protected from peeling reactions by using NaBH₄ as a catalyst in cooking processes (Istek and Ozkan, 2008; Tutus and Cicekler, 2016).

There is little information about the use of Rhododendron species in wood-based industries and there is no study on their use in pulp and paper production. The aim of this study was to evaluate *Rhododendron luteum* and *Rhododendron ponticum* stalks for pulp and paper production, using Kraft-NaBH₄ cooking methods.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

R. luteum and *R. ponticum* species were obtained from Akdamar Village of Akaat district of Trabzon province in Turkey. The chemical compositions of *R. luteum* and *R. ponticum* were determined in a previous study (Birinci, 2008) and are presented in Table 1 with some wood and non-wood species.

According to Table 1, the chemical composition of Rhododendron species is similar to that of hardwoods species. Rhododendron species show similarities among themselves. However, the lignin content of *R. ponticum* was higher than that of *R. luteum*. Due to their high holocellulose contents, it was concluded that Rhododendron species were considered suitable for pulp and paper production.

The samples were chipped and air dried. 12 cooking trials were applied on each species, using Kraft-NaBH₄ method given in Table 2 in order to determine optimum pulping conditions.

Cooking trials of the species were applied in a rotary digester with a 15-liter capacity and high pressure resistant. After cooking processes, the pulps were washed with tap water until the black liquor was removed, and the washed pulps were transferred to 0.15 mm slotted screen. Screened pulp and screen reject yields were calculated after the screening process. The kappa numbers and viscosity of the screened pulps were determined according to TAPPI T 236 om-13 and TAPPI T 230 om-08, respectively (Anonymous, 1998).

The pulps were beaten to 25 ±3 °SR (Schopper Riegler) freeness level in the hollander beater according to TAPPI T 200 sp-96. Ten test papers were produced from the pulps obtained from each cooking trial. The breaking lengths and burst indices of the papers

Table 1 Chemical composition of *R. luteum*, *R. ponticum* and some wood and non-wood species

Tablica 1. Kemijski sastav biljaka *R. luteum*, *R. ponticum* i nekih drvnih i nedravnih vrsta

Chemical components Kemijske sastavnice	<i>R. luteum</i> (Birinci, 2008)	<i>R. ponticum</i> (Birinci, 2008)	<i>A. mem- branaceus</i> (Tutus et al., 2014)	<i>Fagus orientalis</i> (Tank, 1978)	<i>R. pseudoa- cacia</i> (Kirci, 1987)	<i>P. brutia</i> (Tutus et al., 2012)	Wheat straw (Tutus and Cicekler, 2016)
Holocellulose, % holoceluloza, %	78	77	77	79	82	79	73
Alpha cellulose, % alfa-celuloza, %	48	47	50	42	52	49	39
Lignin, % / lignin, %	31	34	24	23	21	28	18
Ash, % / pepeo, %	0.41	0.42	5.50	0.61	0.55	0.48	7.8
Extractives, % ekstraktivi, %	2.02t	2.12t	5.60t	1.50a	6.23a	7.65t	5.1t
%1 NaOH, %	22.2	26.1	29.4	15.6	22.10	14.49	43.7
Hot water, % vruća voda, %	7.15	8.07	8.40	-	-	2.19	14.6
Cold water, % hladna voda, %	1.14	0.88	7.20	1.92	8.06	1.14	11.5

*t – toluene-acetone-ethanol extraction, a – alcohol-benzene extraction / *t – ekstrakcija u toluen-acetonu i etanolu, a – ekstrakcija u alkohol-benzenu

were determined according to TAPPI T 494 om-01 and TAPPI T 403 om-91, respectively (Anonymous, 1998). In order to determine the effects of active alkali and NaBH₄, the optimum cooking parameters were used.

Table 2 Pulping conditions of *R. luteum* and *R. ponticum*

Tablica 2. Uvjeti dobivanja celuloze od biljaka *R. luteum* i *R. ponticum*

Pulping condition Uvjeti proizvodnje celuloze	Unit Jedinica	Value Vrijednost
Active alkali / aktivna lužina	%	18, 20, 22
Sulfidity / sulfidnost	%	24
NaBH ₄ charge / udio NaBH ₄	%	0, 0.3, 0.5, 0.7
Cooking temperature temperatura kuhanja	°C	160
Time to maximum temperature vrijeme postizanja najveće temperature	min	40
Time at maximum temperature vrijeme na najvećoj temperaturi	min	90
Liquor to raw material ratio omjer otapala i sirovine	l/kg	4/1

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

The yield and chemical properties of *R. luteum* and *R. ponticum* pulps used in this study are presented in Table 3.

The screened yield and viscosity of *R. ponticum* was found lower than that of *R. luteum* by about 5.6 % and 15.5 %, respectively. The kappa numbers of *R. ponticum* and *R. luteum* were determined as 49.24 and 45.74. According to these results, *R. luteum* is more suitable for pulping production than *R. ponticum*. The effects of NaBH₄ on the properties of *Rhododendron* pulp are shown in Figure 1.

With the addition of NaBH₄ to the cooking liquor, the yield and viscosity of the pulps increased and kappa numbers decreased. NaBH₄ was effective on the screened yields as it had the ability to stop peeling reactions occurring in cellulose chains (Istek and Ozkan, 2008; Tutus and Cicekler, 2016). NaBH₄ removes the lignin from the pulp i.e. selective lignin delignification occurs, while increasing the viscosity by preserving

Table 3 Screened yields, kappa numbers and viscosities of *R. luteum* and *R. ponticum* pulps

Tablica 3. Prinos prosijavanja, kappa brojevi i viskoznosti celuloze dobivene od biljaka *R. luteum* i *R. ponticum*

Cooking No Broj kuhanja	Active alkali, % Aktivna lužina, %	NaBH ₄ , %	Screened yield, % Prinos prosijavanja, %		Kappa No Kappa broj		Viscosity, cm ³ /g Viskoznost, cm ³ /g	
			<i>R. luteum</i>	<i>R. ponticum</i>	<i>R. luteum</i>	<i>R. ponticum</i>	<i>R. luteum</i>	<i>R. ponticum</i>
1	18	0	42.15	39.78	45.74	49.24	895	832
2	20	0	41.78	38.50	42.16	46.78	860	800
3	22	0	41.05	37.95	41.84	43.18	838	786
4	18	0.3	43.03	41.50	42.06	46.56	928	870
5	20	0.3	42.55	41.00	39.60	44.22	912	854
6	22	0.3	41.83	40.03	37.78	42.76	886	808
7	18	0.5	43.35	41.87	40.10	44.48	949	885
8	20	0.5	42.91	41.21	38.88	41.98	924	868
9	22	0.5	42.00	40.77	36.56	40.06	902	829
10	18	0.7	43.78	42.18	38.98	42.02	988	902
11	20	0.7	43.25	41.65	37.10	39.48	939	880
12	22	0.7	42.60	41.23	34.24	37.98	918	840

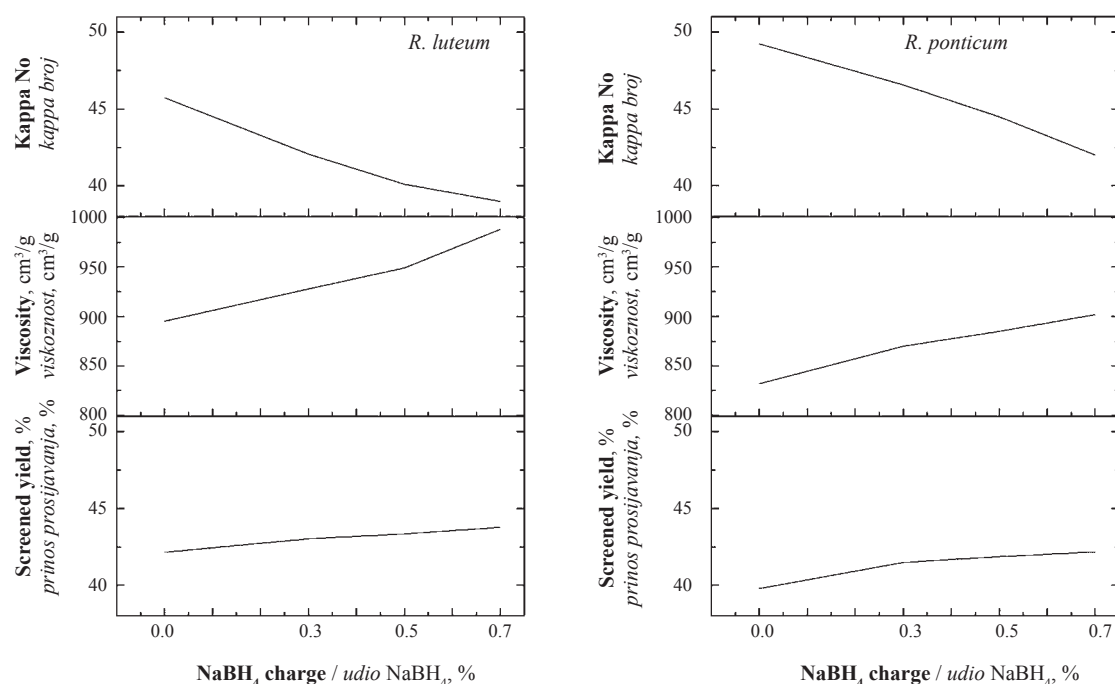


Figure 1 Effects of NaBH₄ charge on yield, kappa number and viscosity of Rhododendron pulps

Slika 1. Utjecaj udjela NaBH₄ na prinos, kappa broj i viskoznost celuloze od rododendrona

carbohydrates (Tutuř, 2008). It also prevents shortening of cellulose and hemicellulose chains as it prevents damage to carbohydrates. In this way, the viscosity of the pulps is increased and strength pulps can be obtained. Recent studies have shown a decrease in the kappa numbers of the pulps obtained with the addition of NaBH₄ to the cooking experiments (Copur and Tozluoglu, 2008; Gulsoy and Eroglu, 2011; Istek and Gonteki, 2009). With the addition of 0.5 % NaBH₄, the screened pulp yields of *R. luteum* and *R. ponticum* pulps increased by approximately 2.8 % (from 42.15 % to 43.35 %) and 5.3 % (from 39.78 % to 41.87 %), while kappa numbers decreased by 12.3 % and 9.8 %, respectively. By keeping the active alkali high, the rate of depolymerization of carbohydrates during cooking increases. Therefore, both yield and viscosity decrease. The screened yields, kappa numbers and viscosity of *R.*

luteum and *R. ponticum* pulps decreased by increasing the active alkali from 18 % to 22 %. Many studies have reported that the increase of active alkali decreases the yield, kappa number and viscosity (Lopez et al., 2000; Yue et al., 2016; Zhai and Zhou, 2014).

The breaking lengths and burst indices of *R. luteum* and *R. ponticum* papers used in this study are given in Table 4.

According to Table 4, the breaking lengths and burst indices of the *R. ponticum* papers were found to be higher than those of *R. luteum*. It is clearly seen that the physical properties of the papers improve with the increase of active alkali and NaBH₄ ratios (Figure 2).

Many studies have reported that boron compounds prevent peeling reaction during cooking, resulting in less damage to carbohydrates and therefore improved physical and optical properties of the papers

Table 4 Breaking lengths and burst indices of *R. luteum* and *R. ponticum* papers

Tablica 4. Duljina lomljenja i indeks pucanja papira proizvedenog od biljaka *R. luteum* i *R. ponticum*

Cooking No Broj kuhanja	Active alkali, % Aktivna lužina, %	NaBH ₄ , %	Breaking length, km Duljina lomljenja, km		Burst index, kPa·m ² ·g ⁻¹ Indeks pucanja, kPa·m ² ·g ⁻¹	
			<i>R. luteum</i>	<i>R. ponticum</i>	<i>R. luteum</i>	<i>R. ponticum</i>
1	18	0	2.21	2.88	1.90	2.10
2	20	0	2.29	2.91	1.98	2.25
3	22	0	2.32	3.02	2.06	2.31
4	18	0.3	2.35	2.98	2.00	2.28
5	20	0.3	2.40	3.05	2.15	2.45
6	22	0.3	2.52	3.09	2.30	2.54
7	18	0.5	2.57	3.07	2.17	2.43
8	20	0.5	2.65	3.10	2.30	2.53
9	22	0.5	2.79	3.19	2.45	2.62
10	18	0.7	2.87	3.15	2.32	2.51
11	20	0.7	3.01	3.32	2.54	2.68
12	22	0.7	3.13	3.51	2.66	2.87

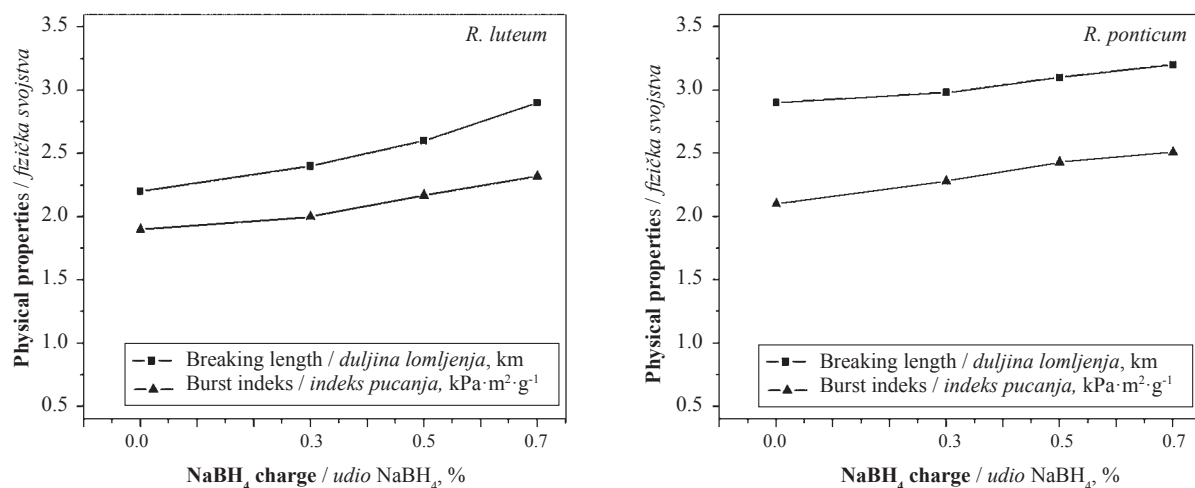


Figure 2 Effects of NaBH₄ charge on breaking length and burst index of Rhododendron papers
Slika 2. Utjecaj udjela NaBH₄ na duljinu lomljenja i indeks pucanja papira od *Rhododendrona*

produced (Akgul *et al.*, 2007; Copur and Tozluoglu, 2008; Istek and Ozkan, 2008; Istek and Gonteki, 2009; Gumuskaya *et al.*, 2011; Erisir *et al.*, 2015; Gulsoy *et al.*, 2016). With the addition of 0.5 % NaBH₄, the breaking lengths of *R. luteum* and *R. ponticum* pulps increased by approximately 16.1 % and 6.4 %, and burst indices also increased by 14.2 % and 15.7 %, respectively. According to data in Tables 3 and 4, optimum cooking conditions for the two species in NaBH₄-added cooking experiments were determined by using 0.5 % NaBH₄ and 18 % active alkali (Cooking No: 7).

4 CONCLUSIONS

4. ZAKLJUČAK

In this study, pulp and papers of *R. luteum* and *R. ponticum* species were produced by using Kraft-NaBH₄ method and their properties were examined. As a result of this study, it was determined that the chemical properties of *R. luteum* and *R. ponticum*, such as yield, kappa and viscosity, were found to be better for pulp and paper industry. Besides, when the physical properties of the produced papers are examined, it is seen that they are suitable for the production of many paper types. The short-fiber pulps obtained from *R. luteum* and *R. ponticum* can be mixed with long-fiber pulps in certain proportions and evaluated for the production of many paper types. Since 70 % of short-fiber raw materials are used for the production of writing-printing paper, the possibility of using *R. luteum* and *R. ponticum* species for the production of different types of quality paper is quite high.

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